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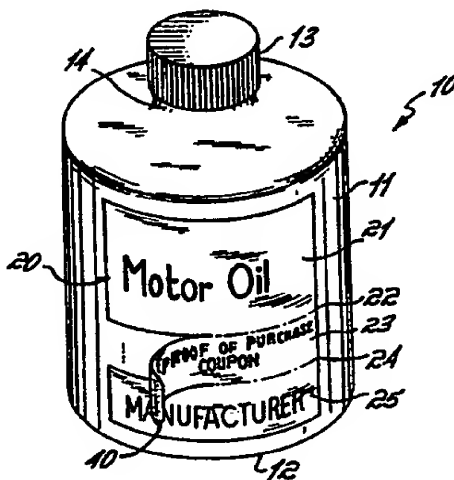
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**(54) Title:** SCORED SYNTHETIC IN-MOLD LABELS AND MOLDED SUBSTRATES LABELED THEREWITH

**(57) Abstract**

Novel, scored synthetic in-mold labels (20) and molded substrates in the form of, for example, hollow plastic containers (10) labeled therewith are disclosed. The novel, scored synthetic in-mold labels (20) are applied to hollow plastic containers via vacuum during the molding operations of the containers. The in-mold labels (20) are prepared from olefin resins and can be in the form of single- or multiple-layered structures. Moreover, the in-mold labels are scored with lines of weakness (22, 24) for dividing the in-mold labels into removable and non-removable sections (23 and 21, 25) which may be separated from one another along the lines of weakness (22, 24) by hand. The non-removable sections (21, 25) can be affixed to the molded containers by permanent adhesives (30) or thermal bonds while the removable sections can be affixed to the molded containers via release coatings, removable adhesives (31) or light thermal bonds. Alternatively, the removable sections (23) may be designed so that they remain unaffixed to the molded containers. The removable sections (23) of the in-mold labels (20) may be in the form of proof-of-purchase coupons.

# + DESIGNATIONS OF "SU"

Any designation of "SU" has effect in the Russian Federation. It is not yet known whether any such designation has effect in other States of the former Soviet Union.

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**Scored Synthetic In-Mold Labels and  
Molded Substrates Labeled Therewith**

**Cross-Reference to Related Applications**

This application for U.S. Patent is a  
Continuation-In-Part of U.S. Patent Application  
assigned Serial No. 07/543,690, filed on June 26,  
5 1990, and entitled "Unstretched Synthetic Papers and  
Methods of Producing Same."

**Field of the Invention**

The present invention relates to synthetic  
10 in-mold labels divided into sections which can be  
separated from one another by hand along score lines  
and which can be applied to molded substrates, such as  
hollow molded containers, during the molding  
operations. The present invention further relates to  
15 molded substrates labeled with such scored synthetic  
in-mold labels.

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Background

Manufacturers of consumer products often attempt to increase their sales by providing labels affixed to their packages which, when removed there-  
5 from, can be redeemed or used to purchase additional like goods at reduced prices. Typically, such labels are prepared from cellulosic base stocks, such as paper, and have proof-of-purchase coupons which can be separated from the rest of the labels by perforations.  
10 The proof-of-purchase coupon areas are generally affixed to the packages so that they can be easily removed therefrom by consumers while the remaining portions of the labels remain affixed to the packages. See, for example, U.S. Patent No. 3,702,511. See also  
15 U.S. Patent No. 4,312,523 and U.S. Patent Re. 30,958.

Today, it is standard practice to manufacture hollow plastic containers by, for instance, blow-molding or injection-molding operations. Generally, the plastic containers formed by means of  
20 blow-molding or injection-molding are fabricated from plastic, polyethylene, polypropylene, polyvinyl chloride, polyethylene terephthalate and the like, and especially high-density polyethylene (HDPE), and are prevalent today in a wide variety of shapes and sizes  
25 for holding many different kinds of materials, such as light duty liquids (e.g., dishwashing detergents),

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heavy duty liquids (e.g., laundry detergents), motor oil, vegetable oil, herbicides, etc.

Generally, the blow-molded or injection-molded containers are provided with a label which  
5 designates the trade name of the product and may contain other information as well. In some instances, the label is merely attached to the container after molding by means of an adhesive or the like. See, for example, U.S. Patent No. 4,587,158 and U.S. Patent Re.  
10 32,929. However, the label may also be attached to the container during the container molding process. This technology by which the label is mounted to the container during the molding operation is generally referred to as an in-mold label (IML) process.

15 In-mold labeling is well known in the art. Generally, an in-mold label is placed in a blow or injection mold prior to the formation of a container. Once the label is in position, the container is formed by blowing or injecting plastic in the mold. The  
20 label, which may include an adhesive, attaches or adheres to the outer surface of the container. In some instances, the label is partially or completely embedded in the plastic surface of the blow- or injection-molded container. The in-mold labeling of  
25 the molded plastic containers allows manufacturers to readily and inexpensively produce labeled containers directly during the molding operations without the

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need to apply labels in subsequent steps. By eliminating subsequent handling, capital investment and handling costs are substantially reduced. See, for example, U.S. Patent Nos. 4,904,324; 4,837,075;  
5 4,605,462; 4,601,926; 4,595,449; 4,323,411; 3,801,689;  
3,292,209; 3,287,198; 3,227,787; 3,207,822; and  
3,108,850.

Notwithstanding the above-mentioned advantages to labeling hollow molded plastic containers  
10 inside the molds, in-mold labels prepared from cellulosic base stocks, such as paper, have proven to be undesirable in operation. For example, it has been found that such conventional paper labels can wrinkle, curl or craze when exposed to heat and moisture  
15 associated with container formation and use. This can result in labels which may not be completely adhered to the outer surfaces of the containers. Furthermore, the wrinkling, curling and crazing of the paper labels provide unsightly aesthetic appearances and detract  
20 from the marketing appeal of the containers labeled therewith. In addition, it has also been found that paper labels associated with certain types of containers, e.g., shampoo containers, which are exposed to very humid environments, e.g., shower stalls, may  
25 eventually disintegrate and become separated from those containers.

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Another problem common with paper-labeled plastic containers is recyclability. Occasionally a defective container will be produced during the molding process. The recycle of scrap molded objects having such labels affixed thereto requires tedious mechanical means or solvents to first remove the labels. In order to reclaim or recycle such plastic containers, particularly defective bottles prepared during the molding process, it is first necessary to remove the paper labels therefrom. Contamination from residual adhesive and small portions of unremoved paper may make it practically impossible to reclaim defective molded plastic containers bearing such labels. Consequently, such defective containers must be burned or buried in land fills, both of which practices are wasteful and undesirable. In view of these drawbacks, the paper label material must be first separated in some manner from the plastic material before the plastic material can be recycled.

Moreover, during the manufacture of hollow molded plastic containers, in-mold labels are generally held in the mold sections by vacuum so that when the semi-molten parisons of plastic material are blown outwardly into conformity with the cavities, or the plastic material is injected into the molds, the in-mold labels become properly adhered or bonded to the hollow molded plastic containers. See, for



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example, U.S. Patent No. 3,759,643 and European Patent No. 281,701 A2. A serious drawback associated with the application of in-mold labels is that, if they are perforated, they will not be held properly, if held at all, in the mold sections by the vacuum during the molding operations. Consequently, in-mold labels provided with, for example, tearable proof-of-purchase coupons have been commercially unavailable hitherto.

Accordingly, there is a demand in the industry for synthetic in-mold labels which are divided into sections which can be torn by hand along lines of weakness for removing sections in the form of, for example, proof-of-purchase coupons, from hollow plastic containers after the containers have been labeled with such in-mold labels during the molding operations.

#### Summary of the Invention

In brief, the present invention alleviates certain of the above-mentioned disadvantages and shortcomings of the present state of the in-mold label art through the discovery of novel, scored synthetic in-mold labels which are tearable by hand along lines of weakness and molded substrate labeled therewith. Generally speaking, the in-mold labels of the instant invention are in the form of single- or multi-layered plastic films, which are intended to be applied to

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molded substrates, such as hollow molded containers, during the molding operations. The novel in-mold labels are typically prepared from olefin resins and are scored with lines of weakness for dividing the in-mold labels into unique removable and non-removable sections which may be separated from one another by hand. The non-removable sections of the in-mold labels of the instant invention are affixed to the molded substrates by permanent bonding means, such as permanent adhesives or thermal bonds, while the removable sections of the same in-mold labels are affixed to the molded substrates via removable bonding means, such as release coatings, removable adhesives or light thermal bonds. Nevertheless, in some instances, depending upon the materials selected to manufacture the molded substrates and the in-mold labels of the instant invention, the removable sections of the in-mold labels may remain unaffixed to the molded substrates so long as the other sections thereof are permanently bonded to the molded substrates. The molded substrates are typically prepared from olefin resins and can be produced by for instance blow molding or injection molding techniques or the like.

In accordance with the instant invention, the novel synthetic in-mold labels include score lines which function to divide the synthetic in-mold labels into sections which can be easily separated from one

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another by hand along the score lines when the areas encompassing the score lines are subjected to sufficient non-parallel stresses. In other words, the synthetic in-mold labels are provided with fine, unperforated score lines whereby the labels may be torn by hand along the score lines to remove sections therefrom. Moreover, because the score lines may be continuous or interrupted, but not perforated, the novel in-mold labels of the instant invention can be uniquely applied in the molds via vacuum to the molded containers during the molding operations.

The score lines for dividing the novel synthetic in-mold labels of the instant invention into sections can be formed by, for example, engaging a sharp blade, such as a razor blade or a laser beam, into the surface of the in-mold label material at a selected or predetermined depth as the material passes over a smooth roller or stationary bar. This can be accomplished, for instance, by applying a selected force to the sharp blade through the use of an air cylinder, a weight, a spring, an electromagnet or the like, or by establishing a blade protrusion beyond a fixed wide shoulder and applying the wide shoulder to the surface of the material being scored by the application of force. Thus, it should be readily apparent that the novel synthetic in-mold labels of the instant invention can be amazingly applied to

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molded containers during the molding operations and constructed with removable sections, such as proof-of-purchase coupons or the like, which can be easily torn by hand along the score lines from the remainder  
5 of the in-mold label sections affixed to the molded containers.

While the in-mold labels of the present invention can be stretched or unstretched and made from many suitable polymers, the novel synthetic  
10 in-mold labels of the instant invention are preferably unstretched, i.e., substantially free of mechanically produced microvoids, and are formed with a continuous polyolefin resin matrix containing an effective amount of a particulate filler having pre-existing microvoids  
15 provided therein. The microvoid particulate fillers of such unstretched in-mold labels are dispersed throughout the continuous olefin resin matrix of the synthetic in-mold labels of the instant invention to provide microvoids in communication with the surfaces  
20 of the synthetic in-mold labels for imparting thereto receptivity and retentivity for the common writing and printing materials. Such synthetic unstretched in-mold labels according to the present invention are believed to have excellent stiffness, tensile  
25 strength, and flexibility as are required for paper materials, they are generally uniform in thickness, and they have good tear resistance and writable and

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printable properties. These surprising characteristics are believed to incur since the microvoid particulate fillers, which are integrally formed with the continuous olefin resin matrix, are dispersed somewhat uniformly but randomly therethroughout so that the non-mechanically produced microvoids are provided at the exterior surfaces of the in-mold labels to render such surfaces porous for imparting writability and printability thereto. Moreover, the surprising writable and printable characteristics are amazingly accomplished with the preferred in-mold labels of the instant invention even though they are not mechanically stretched to generate mechanically produced microvoids.

A method for producing preferred in-mold labels according to the instant invention provides for admixing the constituents into, for example, a homogeneous blend in the form of a pellet and extruding the pellet into a polymer matrix in the form of a film or sheet having a thickness (gauge) of about 1 mil to about 10 mils or more and preferably of about 4 mils to about 6 mils for making the in-mold labels. The preferred polymer resins which can be used include, for instance, polyethylene, polypropylene, copolymers and the like. Microvoid particulate fillers which are suitable for use include, for example, diatomaceous earth, volcanic ash, silica gels, styrogels, porous

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glass beads or other such equivalents. The polymer matrix may also include other additives for achieving desired properties. For example, the polymer matrix of the synthetic in-mold labels of the instant invention may include a foldability improving agent, such as a styrene resin, in a specific content. By introducing the styrene resin into the polymer matrix, the foldability of the instant synthetic in-mold labels is believed to be improved. Moreover, by the addition of a styrene resin into the polymer matrix, it is believed that the synthetic in-mold labels of the present invention acquire the appearance of being multi-layered which is commonly associated with conventional pulp writing paper.

The unique and surprising writable, printable and paper-like properties associated with the preferred synthetic in-mold labels of the present invention are amazingly accomplished without having to resort to other complex and/or expensive mechanical, chemical and electrical processes. Accordingly, when following the teachings of the instant invention, manufacturers are not required to mechanically stretch the preferred synthetic in-mold labels to generate mechanically produced microvoids for imparting writability and printability thereto, since certain synthetic in-mold labels of the instant invention can be formulated with a continuous polymer matrix which

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includes an effective amount of a particular filler having pre-existing microvoids formed therein for providing microvoids at, on or near the exterior surfaces thereof for imparting writability and printability thereto. Nevertheless, if desired, the synthetic in-mold labels of the instant invention may be formulated with other polymers or additives, or coextruded with, for example, polystyrene or the like to impart further writability to the exterior surfaces thereof. Moreover, if desired, the synthetic in-mold labels of the instant invention may be electrically, mechanically or chemically treated to also further impart writeability to the exterior surfaces thereof so long as the scoring objectives of the present invention are not defeated.

The present invention therefore now makes it possible to produce synthetic in-mold labels for hollow molded containers which can be applied during the molding operations and which include removable sections, such as proof-of-purchase coupons, which can be torn from the labels positioned on the molded containers by consumers along tear lines for redemption or use to purchase additional like goods at reduced prices. Moreover, the novel synthetic, sectionalized in-mold labels of the instant invention uniquely afford cost saving and convenient advantages by permitting the plastic hollow molded containers

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labeled with such in-mold labels to be easily recycled without having to first remove the labels therefrom. It should therefore now be apparent to those versed in this field that certain of the problems attendant with paper and/or plastics in-mold labels provided hitherto  
5 are overcome in a relatively simple, yet unobvious manner by the present invention.

The above features and advantages of the instant invention will be better understood with  
10 reference to the accompanying figures, detailed description and examples. It should also be understood that the particular synthetic in-mold label illustrating the instant invention is exemplary only and not to be regarded as a limitation of the inven-  
15 tion.

#### Brief Description of the Figs.

Reference is now made to the accompanying Figs. in which certain Figs. illustrate embodiments of the present invention from which its novel features  
20 and advantages will be apparent:

Fig. 1 is a view magnified by 5,000X of diatomaceous earth particulate filler particles having pre-existing microvoids inherent thereto which can be  
25 utilized in the instant invention;

Fig. 2 is a view magnified by 5,000X of a portion of a surface of an unstretched synthetic



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paper-like film produced with diatomaceous earth particulate filler particles having pre-existing microvoids inherent thereto in accordance with the instant invention, which illustrates such a particle  
5 on or at the surface thereof for imparting write-ability and printability thereto;

Fig. 3 is a view magnified by 5,000X of a portion of a cross-section of an unstretched synthetic paper-like film produced in accordance with the  
10 instant invention illustrating diatomaceous earth particulate filler particles having pre-existing microvoids inherent thereto which are dispersed therethroughout, as indicated by a sponge-like texture;

15 Fig. 4 is a view magnified by 5,000X of calcium carbonate particles which appear to be devoid of any pre-existing microvoids;

Fig. 5 is a view magnified by 5,000X of a portion of a surface of an unstretched plastic film  
20 formed with calcium carbonate particles devoid of microvoids, which illustrates such a particle on or at the surface thereof;

Fig. 6 is a view magnified by 5,000X of a portion of a cross-section of an unstretched plastic  
25 film formed with calcium carbonate particles devoid of microvoids which are dispersed therethroughout;

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Fig. 7 is a photomicrograph of a cross section magnified approximately 560X of an in-mold label of the instant invention which is approximately 0.004 inches thick with a score depth of approximately 33% of the thickness;

Fig. 8 is a graph of the tear strength of a score line versus the depth of a score line as a percentage of material thickness for an in-mold label formulated with a composition designated as composition I in Example I;

Fig. 9 is a rear plan view of a plastic in-mold label of the present invention;

Fig. 10 is an enlarged fragmentary sectional view of the in-mold label shown in Figs. 9 and 11; and

Fig. 11 is a perspective view of a hollow container labeled with the plastic in-mold label shown in Figs. 9 and 10.

#### Detailed Description

By way of illustrating and providing a more complete appreciation of the present invention and many of the attendant advantages thereof, the following detailed description is given concerning the novel plastic in-mold labels and molded substrates, such as hollow molded containers, labeled therewith.

The scored synthetic in-mold labels of the instant invention may comprise one or more layers and

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are intended to be applied to molded substrates, such as hollow molded containers, typically made of olefin plastic materials during the molding operations. The scored in-mold labels of the present invention are also typically made of olefin plastic materials.

Referring now to Fig. 11, a hollow plastic container 10, such as a plastic container for motor oil, comprising an outer surface 11, a bottom wall 12, and a cap 13 mounted on a neck 14 is shown. In Fig. 11, hollow plastic container 10 is depicted with a sectionalized in-mold label 20 affixed thereto. Sectionalized in-mold label 20 as shown in Fig. 11 comprises permanent sections 21 and 25 divided from removable section 23 via score lines 22 and 24, respectively. As further shown in Fig. 11, removable section 23 is provided with edge tab 40 for facilitating removal of removable section 23 along score lines 22 and 24 from permanent sections 21 and 25 and from the outer surface 11 of hollow plastic container 10. With reference to Figs. 9 and 10, permanent adhesive 30 and removable adhesive 31 are shown on the inner surfaces of permanent sections 21 and 25 and removable section 23, respectively. A hollow plastic container labeled with an in-mold label of the instant invention is made by, for example, conventional blow molding techniques wherein a preform or parison at the blow molding temperature is enclosed in a hollow mold

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which has an in-mold label suitably held against a surface of the cavity of the mold by, for example, vacuum, and the parison is expanded or blown outwardly against the confines of the mold so that the in-mold label is affixed to the outer surface of the resultant hollow plastic container.

In Figs. 9-11, in-mold label 20 is made from an olefin plastic material in the form of a plastic film layer having a surface which is adapted for receiving and retaining thereon common writing and printing materials and having permanent sections 21 and 25 separated from removable section 23, which can be in the form of a proof-of-purchase coupon, by score lines 22 and 24, as indicated above. By the term "score line(s)," it is meant herein to refer to a line of relative weakness in the form of a groove along a predetermined path in one or more layers of an in-mold label of the instant invention which allows the in-mold label to be preferentially folded, torn, separated, divided or otherwise manipulated along that path in a manner that was not possible without the line of relative weakness, particularly when the area encompassing the line of relative weakness is subjected to sufficient stresses which are non-parallel thereto. By the term "groove" it is used herein to refer to any alteration that penetrates only a portion of but does not cut through the total thickness of an

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in-mold label of the instant invention. Accordingly, for the purpose of the present invention, the term "score line(s)" as used herein excludes perforations.

It is presently believed that the lines of relative weakness imparted to the in-mold labels of the present invention are typically a result of two factors. These are a reduction in cross-sectional thickness of the material selected to form the in-mold labels of the instant invention and an introduction of a flaw or stress riser therein, e.g., the grooves. The relative effectiveness of these factors in causing the tear or separation to propagate along the score line of course depends primarily on the nature of the material selected to form the in-mold labels. Typically, a synthetic film selected to form an in-mold label of the instant invention having thickness ranges and compositions disclosed herein may have a groove depth of about 10% to about 50% of the film thickness (gauge) with a preferred depth of about 33%, as illustrated in Figs. 7 and 8. With reference to Fig. 8, it can be seen that when the groove depth is less than about 10% of the material thickness, the tear strength is believed to be too great whereas when the groove depth is greater than about 50% of the material thickness, the material strength is believed to be rendered too weak to operate in accordance with the present invention. Notwithstanding, it is to be

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understood that values outside of these ranges may produce workable score lines for these or other materials and applications, but the stated ranges are believed to produce a good compromise between basic material strength properties and consistent functionality of the score lines in facilitating clear separation of the material. Thus, the depth of a groove in a synthetic film selected to form an in-mold label of the instant invention can conceivably be of any depth so long as the depth of the groove is sufficient to accomplish the objectives of the instant invention.

Although it should be understood that other methods may be employed to produce a score line or a plurality of score lines, the following represents a workable approach. It is presently believed that this method is well suited for producing machine direction score lines in substantially continuous web processes. Basically, the method involves the engagement of a sharp blade, such as a razor blade, or laser beam, into the surface of a moving web as it passes over a smooth back-up roller or stationary bar. The blade is engaged by application of a force through the use of, for example, an air cylinder, weight, spring, electro-magnet or the like. The required depth of cut is achieved by adjusting the force, typically in a range of between about 0.1 and about 3 lbs. The necessary

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force of course is a function of depth of cut, blade sharpness, material properties, back-up roller diameter, and other factors.

As an alternative method, groove depth can  
5 be regulated by establishing a blade protrusion beyond  
a fixed wide shoulder. The shoulder would be pressed  
into contact with the surface of the material being  
grooved by the application of a force. The blade  
would then engage the surface of the material by the  
10 amount of a preset protrusion. For example, a blade  
protrusion could be preset for about 0.001" for  
grooving a material of this invention to produce a  
groove depth of approximately 0.001".

It should be further understood that line  
15 speed of the process affects the quality of the score  
line. The grooving process as disclosed is believed  
to have no lower speed limit, and the upper speed  
limit has not been determined since it is believed to  
be outside of the range of speeds normally used to  
20 process materials of the invention. Depending upon  
the material selected, blade sharpness and other  
factors, the score lines formed tend to become more  
easily detectable as line speed is increased. This is  
believed to be due to localized heating which softens  
25 the material, allowing it to be displaced out of the  
groove areas creating a furrow effect on the surface  
of the material along the score lines. In most

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product applications, this microscopic roughness surrounding the score lines, as shown in Fig. 7, is of no significance. In the in-mold label application described herein, it is believed that score lines can be formed on either side of the film and at over 300 feet per minute with the score lines being virtually invisible on the final products. As indicated hereinabove, the score lines of the instant invention may be formed by any other suitable methods, such as those disclosed in U.S. Patents, No. 4,217,327, No. 3,379,814, No. 3,909,582, and No. 3,292,513, all of which are incorporated herein by reference in their entireties, so long as the objectives of the instant invention are not defeated. In addition, while the Figs. disclose an in-mold label having two parallel score lines, it should be understood that the in-mold labels of the present invention can be formed with one score line or any number of score lines in a parallel or non-parallel relationship for carrying out the objectives of the instant invention.

Referring again to Figs. 9-11, permanent sections 21 and 25 of in-mold label are permanently bonded to the outer surface 11 of the hollow plastic container 10 via permanent adhesive 30 whereas removable section 23 is removably bonded to the outer surface 11 of the hollow plastic container 10 via removable adhesive 31. Exemplary of permanent adhe-



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sives that may be employed include adhesive No. CA-231 distributed by Century Adhesives, adhesive No. J9775D (EVA gel lacquer adhesive) distributed by Pierce and Stevens, and adhesive No. 4983 distributed by

5 Michelman. Exemplary of removable adhesives that may be employed include adhesives distributed by Air Products under the names AIRFLEX 400 and AIRFLEX 465 (very slightly tacky), and by Unocal under product No. 46130 (slightly tacky). Exemplary of other potential

10 adhesives include those disclosed by Wiesman, D.K. et al., Water-Borne Heat Seal Coatings for In-Mold Labeling, TAPPI Proceedings, pp 29-33 (1990 Polymers, Laminations & Coatings Conference), such as the solvent-borne gel lacquer heat seal coatings, the

15 water-borne heat seal coatings and the 100% solids heat seal coatings, which cited disclosure is incorporated herein by reference in its entirety. Examples of still other potential adhesives include those set forth in for example U.S. Patents, No. 4,837,075, No.

20 4,601,926 and No. 4,587,158, and European Patent No. 281,701 A2, all of which are incorporated herein by reference in their entireties.

For a film of the instant invention having a dry film thickness of about 0.75 mils and a composition recited herein and in particular in Example I,

25 such adhesives can be cast from a water emulsion onto the inner surface of the film at an amount of about 15

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gm/yd<sup>2</sup> or less and dried for a suitable time at a suitable temperature known to those versed in the adhesive art. The application roll for the Pierce and Stevens' adhesive No. J9775D may be a 140-150 line quad application roll which, when applied via such roll at that amount, can be dried at about 140-150°F. Once dried, the laid down adhesive can be heat sealed to the inner surface 11 of the hollow plastic container 10 during the blow- or injection-molding processes. It is believed that when the Michelman 4983 adhesive is laid down in the stated amount on a film having the composition recited in Example I, such adhesive has an adhesive strength of about 5 pli whereas the AIRFLEX 400, AIRFLEX 465 and Unocal 6130 adhesives are believed to each have an adhesive strength of about 0.15 pli, about 0.50 pli and about 0.50 pli, respectively, on the same film when laid down in about the same amounts. Generally speaking, it should be understood that the adhesive strength for the removable adhesive should be preferably less than or equal to about 0.2 pli. Thus, the AIRFLEX 465 and Unocal 6130 adhesives are believed to be less desirable at the above-stated lay down amounts. Moreover, because these two adhesives are somewhat tacky, they may be less desirable than the AIRFLEX 400 adhesive for reasons discussed hereinbelow.

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It should of course be understood that any suitable adhesive which permits the permanent sections of an in-mold label of the instant invention to remain bonded to the outer surface 11 of hollow plastic container 10 or permits the removable sections to be removed therefrom are contemplated by the instant invention. Thus, while the terms "permanent" and "removable" are used herein in connection with bonding various sections of the in-mold labels of the instant invention to substrates such as plastic molded containers, it should be understood that these terms are being used in a relative, rather than absolute sense. It should be further understood, however, that the actual lay down amounts and drying times for any adhesive may vary and will depend upon the particular adhesive selected as well as the objectives intended therefor. Moreover, it should be appreciated that when a tacky or pressure sensitive adhesive is selected, there is the possibility that it may interfere with the label making operations concerning the steps involving sheet and die cutting the in-mold labels into stacks. Such tacky adhesives can also interfere when attempting to separate individual in-mold labels from the stacks for insertion into the molds during the molding operations. It should be further understood that the permanent and removable bonding means are preferably located on those surfaces

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of the in-mold labels which make contact with the molded substrates during the molding operations. Thus, when the permanent and removable bonding means are permanent and light thermal bonds, respectively, the permanent and removable bonding means are the surfaces of the in-mold labels making contact with the molded substrates, whereas when the permanent and removable bonding means are permanent and removable adhesives, respectively, such adhesives are positioned on the surfaces of the in-mold labels making contact with the molded substrates.

Referring now to Fig. 11, the outer surface of in-mold label 20 is typically formed with suitable indicia or graphics such as printing to produce identification as well as desired aesthetic effects. Thus, in-mold label 20 can be provided with normal trademark and instructional material printed thereon in the permanent as well as removable sections, as shown in Fig. 11. In addition, if desired, the in-mold labels of the instant invention may have printed matter on one or both sides thereof. Thus, the removable section 23 of in-mold label 20 may be used for promotional purposes as a proof-of-purchase label. The printing on the outside of the removable section 23 may contain instructions, for example, on how to redeem the proof of purchase label or to remove the proof-of-purchase label without destroying it.

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In-mold label 20 is applied to the hollow plastic container 10 to form a unit as shown in Fig.

11. In one embodiment, in-mold label 20 has a permanent adhesive 30 on the back side of portions 21 and  
5 25 and a removable adhesive 31 on the back side of removable section 23 thereof, as indicated hereinbefore. In another embodiment, the permanent sections 21 and 25 of in-mold label 20 can be formed with an olefin plastic material or layer which is compatible  
10 with the outer surface 11 of hollow plastic container 10 so that during the molding process, these permanent sections are heat bonded to outer surface 11. In this embodiment, the use of a foamed polystyrene label stock, such as Opticite<sup>TM</sup> distributed by Dow Chemical,  
15 or a coextruded label stock with an ethylene-vinylacetate (EVA) layer contacting the container could be used. In addition, it is believed that bonding could be enhanced by the use of corona treatment and high parison temperature.

20 As an alternative to a removable adhesive, removable section 23 can be formed with an olefin plastic material or layer which is not compatible with outer surface 11 of the hollow plastic container 10 or with a thermally insulative layer so that removable  
25 section 23 will form either a light thermal bond or no thermal bond at all during the molding process. It is believed that this level of bonding can be tailored

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through control of temperature of the parison, temperature of the mold, molding pressure and/or, as already indicated, label composition.

In yet another alternative to the removable adhesive, removable section 23 can be bonded to the outer surface 11 of the hollow plastic container 10 via a release coating. To achieve a removable bond under this embodiment under conditions where a permanent thermal bond would otherwise have been produced, a release coating formed of, for example, a silicone, paraffin, lacquer or the like, could be applied in a line or dot pattern of any geometric type. Of course, it should be understood that the larger percentage of area covered by the release coating, the lower the remaining peel strength will be.

In another embodiment for affixing the in-mold labels of the instant invention to the hollow plastic containers, permanent adhesive bonding can be utilized only in the permanent sections in those cases where little or no thermal bonding is achieved. If desired, the same permanent adhesive may be applied to the removable section of the in-mold label, but in a line or dot pattern of any geometric type to reduce the percentage of surface area bonded to effect a removable bond between the removable section of the in-mold label and the hollow plastic container. Thus,

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it should be readily apparent that these or any combinations involving thermal bonds, adhesives and release coatings may be employed to affix the in-mold labels of the instant invention to the outer surfaces of the hollow plastic containers during the molding process so that the objectives of the instant invention may be carried out.

Also, if desired, removable section 23 may be provided with an edge tab 40 for facilitating the removal of removable section 23 from the hollow plastic container 10, as shown in Figs. 9 and 11 and indicated hereinbefore. Of course, it should be understood that when the in-mold labels of the instant invention are formed with such edge tabs, the inner surfaces of the edge tabs should be formed with an incompatible olefin plastic layer or a thermally insulative layer so that either a very light thermal bond or no thermal bond will result during the molding process. If the olefin plastic selected to formulate the in-mold labels of the instant invention does not form thermal bonds with the outer surface of the hollow plastic containers during the molding operations, no further treatment of the edge tabs is required. As an alternative, the adhesive selected for the removable section of the in-mold label or other adhesives may be laid down over the edge tab area in the same or lesser amounts for forming a

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removal bond between the edge tab and outer surface of the hollow plastic container, as shown in Fig. 9.

The plastic in-mold labels of the instant invention are preferably produced by a process comprising melt mixing and forming a homogeneous blend comprising an olefin resin in an amount by weight of about 60% to about 90% and a particulate filler having pre-existing microvoids provided therewith in an amount by weight of about 10% to about 40%, and thereafter shaping the melt mixed homogeneous blend into a film or sheet having a continuous matrix. The microvoid particulate filler is dispersed somewhat uniformly but randomly throughout the continuous matrix such that microvoids are provided in communication with the surface for imparting writeability and printability thereto, as illustrated in Figs. 1-3. Once produced, the film or sheet can be scored, written or printed on, adhesive or release coated on one side thereof if desired, and die cut to form the in-mold labels.

By the phrases "particulate filler having microvoids" or "microvoid particulate filler," they are used in a broad sense herein and refer to any organic or inorganic substance in the form of particles having microvoids for imparting porosity, such that when the particulate filler is blended into the continuous olefin resin matrix, the particulate filler



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provides microvoids at, on and/or near the surfaces of the unstretched synthetic in-mold labels produced to impart writeability and printability thereto, as depicted in Figs. 1-3 and 8. Nevertheless, it should be understood that the terms "particulate filler having microvoids" or "microvoid particulate filler" are also intended to include any organic or inorganic substance which has the ability to impart effective porosity to at least the surface of an unstretched synthetic in-mold label produced in accordance with the teachings of the instant invention to impart acceptable writeability and printability thereto. The terms "particulate filler having microvoids" or "microvoid particulate filler" are not meant, however, to include those fillers which are incapable of imparting porosity to the surface of the in-mold labels produced for imparting good writeability or printability thereto, such as particulate fillers virtually devoid of microvoids and formed of nonporous materials, like calcium carbonate, as illustrated by Figs. 4-6.

By the term "microvoid" as used herein in the above-phrases, it too is used in a broad sense herein and refers to voids smaller than about 20 microns, but nevertheless of any sufficient size for receiving and retaining writing materials for imparting writeability to the surfaces of the in-mold labels

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of the present invention. See for example the particulate filler depicted in Fig. 1, as compared to the particles illustrated in Fig. 4.

Examples of microvoid particulate fillers that can be used in the instant invention include diatomaceous earths, volcanic ash, silica gels, styrogels, porous glass beads and the like. Examples of diatomaceous earths that may be dispersed within the continuous olefin resin matrix of the plastic in-mold labels of the instant invention include Eagle-Picher's Celaton MW-21, MW-25 and MW-27, Witco's Micro-Ken 801, and John Manville's Celite 212. An example of volcanic ash that may be utilized with the present invention is Grefco Inc.'s Perlites material, such as FF16 and FF26. Regardless of which microvoid particulate filler is selected, it should be understood that the microvoid particulate filler should have the properties defined above so that when dispersed throughout the continuous olefin resin matrix of the in-mold labels in an effective amount, an effective amount of microvoids are in communication with the surface to impart writeability and printability thereto. The amount of microvoid particulate filler to be used will of course be influenced by the size of the particles, the type of olefin resin selected, the amount of other additives used, the molding conditions, the thickness of the in-mold

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labels produced and the like, but generally, such particulate fillers are used in an amount of between about 5% and about 30% by weight. Typically, the microvoid particulate fillers have a particle size in the range of between about 2 microns and about 20 microns, and preferably between about 5 microns and about 10 microns.

The olefin resins that can be used to formulate the in-mold labels of the instant invention are, for example, high density polyethylene, medium density polyethylene, low density polyethylene, ethylene-vinylacetate copolymers, ethylene-propylene copolymers, polypropylene, polybutene-1 and the like. Such olefin resins may be used individually, or in combination. High density polyethylene is especially preferred and examples thereof include those available through Dow Chemicals, HDPE (10062) and Quantum Chemical HDPE (6206).

According to the present invention, it may also be desirable to add other additive resins to the above-mentioned composition. For instance, when a styrene resin or the like is used as the additive resin, good printability, stiffness as well as foldability is imparted to the in-mold labels. It is believed, however, that good foldability typically is not obtained with a styrene resin content of less than about 5% by weight. On the other hand, it is believed

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that the styrene resin content should not be greater than about 10% by weight in order to prevent substantial lowering of the tear strength, impact strength and tensile strength of the in-mold labels produced therewith. Examples of styrene resins that can be used in this invention include polystyrene, poly-alpha-methylstyrene, styrene-butadiene copolymers having a high styrene content, high impact polystyrene, acrylonitrile-butadiene-styrene copolymer, acrylonitrile-styrene copolymer, styrene-methylene-methacryate copolymer and the like. When copolymers of styrene with other monomers are selected, it is believed that the styrene content should be at least about 50%. An example of a polystyrene resin that can be used in the instant invention may be obtained from Dow Chemicals under the trademark Styron 420 or from Phillip under polystyrene resin.

In this invention, an inorganic filler may be further added to the in-mold label mixture of the olefin resin and microvoid particulate filler with or without the above-mentioned additive resins, such as a styrene resin. As to the inorganic filler, talc, kaline, zeolite, mica powder, asbestos powder, calcium carbonate, magnesium carbonate, calcium sulfate, clay, silica powder, aluminum magnesium sulfate, barium sulfate, zinc sulfite, titanium dioxide, zinc oxide and the like may be used. When an inorganic filler is

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selected, it is preferred that such inorganic filler have an average particle size of less than about 10 microns, and more preferably less than about 5 microns. The inorganic fillers can be used in amounts of between about 0 and about 20% by weight, and preferably between about 5% to about 10% by weight. Among such inorganic fillers, calcium carbonate is particularly preferred and may be obtained from ECCA Calcium Products, Inc., Supercoat, Georgia Marble, Gamma Sphere  $\text{CaCO}_3$ , and Pfizer, multi-flex MM coated and uncoated  $\text{CaCO}_3$ . While calcium carbonate is a preferred inorganic filler, it should be appreciated that it is not to be used as a substitute for the microvoid particulate filler to impart writeability to the surfaces of the in-mold labels of the instant invention when they are not stretched since calcium carbonate particles are generally devoid of the necessary microvoids, as illustrated by Figs. 1-6. Of course, it should be understood that, when it is desirable to stretch the in-mold labels of the instant invention, microvoid-free inorganic fillers, such as calcium carbonate, may be used with or substitutes for the particulate fillers having microvoids since effective stretching of the in-mold labels will impart the necessary porosity to the exterior surfaces thereof for permitting writing and printing thereon.

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For the purpose of improving moldability of the composition and physical properties of the resultant in-mold labels and increasing the amount of the particulate and/or inorganic filler in the composition, it is believed that natural rubber or synthetic rubber such as polyurethane rubber, styrene-butadiene rubber, acrylonitrile-butadiene rubber, poly-butadiene rubber, polypropyleneoxide rubber and the like may be added to the continuous olefin resin matrix. Such rubber, however, should not be used in an amount which exceeds the total amount of the olefin resin, any additive resin, such as a styrene resin, and/or the particulate filler. If an additive resin already containing rubber, such as high impact polystyrene, is used, this should be considered when adding other rubber containing materials to the composition.

In the present invention, the in-mold labels may be pigmented to impart desirable aesthetic qualities by for example incorporating into the continuous matrix a colored or white pigment, such as titanium dioxide. In addition to being pigmented, it is also possible to further add other various additives to the in-mold labels such as a plasticizer, a stabilizer or other similar agents. Further, for promoting the dispersion of the particulate filler and/or inorganic filler, it may be advantageous to use a surfactant, a

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dispersing agent or other similar agent. It is also possible to add an ultraviolet ray absorbent or an anti-oxidant so as to improve weatherability of the resultant structure, or to add an anti-static agent.

5 These types of additives would be employed in their customary amounts well known to those versed in this field.

When a product of the present invention is produced as described above, an in-mold label having

10 the appearance of multi-layers may result due to the fact that when an additive resin is blended into the composition, the resultant product has a tendency of sometimes undergoing separation of the olefin resin from the additive resin, such as a styrene resin, to

15 create the appearance of a multi-layered-like structure which is characteristic of conventional pulp paper. As a result, an in-mold label produced in accordance with the instant invention has the characteristics of being stiff and foldable, and it can be

20 torn. In addition, when a styrene resin is utilized, it is believed that various layers may be peeled from the in-mold labels produced in accordance with the instant invention which is further characteristic of conventional pulp paper.

25 The above-mentioned specific compositions are typically melt mixed sufficiently by means of, for example, a mixing roll, a Banbury mixer, an extruder

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or the like, and then molded into a film or sheet by means of, for example, a calendar roll, an extruder or the like. Generally, the molding is performed in a manner such that the resultant film or sheet product  
5 has a continuous olefin resin matrix with a thickness (gauge) of between about 1 mil and about 10 mils, with a preferable thickness of between about 4 mils and about 6 mils.

The present invention also admits of a  
10 number of variations all within the scope thereof. For example, it is possible to co-extrude the preferred polymer material with a sheet of other material to form a multi-layered plastic film layer. One possibility is to form a sheet of tearable decorative  
15 material 6 mils in thickness by co-extrusion of 4 mils of the preferred compositions set forth above and 2 mils of high density polyethylene. The co-extruded sheet may then be embossed or otherwise treated to form the desired tear lines before undergoing further  
20 processing to form the in-mold labels. Another possibility is to form a plastic film layer comprising a sheet of tearable decorative material 6 mils in thickness by co-extrusion of 4 mils of a suitable synthetic material and 2 mils of polystyrene to impart  
25 writeability and printability thereto. Yet another possibility is to form a plastic film layer by extrusion coating the preferred polymeric film material on



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paper, scrim or other substrate. Of course, if a paper substrate is selected, the resultant in-mold labels may defeat the recycle advantages associated with all plastic in-mold labels. Examples of additional synthetic labels that may be employed in accordance with the teachings of the present invention include those disclosed in U.S. Patents, No. 4,904,324, No. 4,837,075 and No. 4,601,926 and in European Patent 281,701 A2, all of which are incorporated herein by reference.

The in-mold labels of the instant invention are typically first formed into films or sheets which can be scored, adhesive or release coated, printed and die cut into plastic in-mold labels for labeling plastic molded articles, such as bottles, produced by, for example, blow-molding or injection-molding operations. The plastic in-mold labels of the instant invention have many advantages over paper labels. For example, the plastic in-mold labels and plastic articles to which they are applied can be easily and advantageously recycled. Still further, the in-mold labels can be formed with removable sections, such as proof-of-purchase coupons or the like, which can be torn from the main portions of the in-mold labels by hand along the score lines without significant loss of tensile strength of the in-mold labels as a whole or without removing the main portions of the in-mold

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labels affixed to the molded containers. Moreover, the in-mold labels of the present invention may be applied to the plastic molded containers by vacuum during article molding operations in accordance with techniques well known to those versed in the art, such as those described in U.S. Patents, No. 4,708,630, No. 3,108,850, and No. 3,417,175, all of which are incorporated herein by reference in their entireties.

A scored, synthetic in-mold label of the instant invention and a hollow molded plastic container labeled therewith can be produced in accordance with the following brief description. A roll of film produced in accordance with Example I is unwound and past through scoring equipment. All score lines, which are preferentially made simultaneously, are positioned relative to one another and a reference edge. Once the score lines have been made, the film is rewound. The scored roll of film is then unwound into appropriate adhesive coatings and printing equipment. The adhesive coats are patterned relative to a reference edge with three levels of adhesive being laid down on preferably the score line side. A high level of adhesive is laid down where the removable section, i.e., proof-of-purchase coupon, is not. A low level of adhesive is laid down where the removable section, i.e., proof-of-purchase coupon, is. With respect to the edge tab, no adhesive is laid down

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at the edge tab portion of the removable section. If desired, however, the same low level of adhesive that is laid down over the removable section can be laid down over the edge tab region in the same or lesser amounts. Once the adhesives have been properly laid down, the solvents are flashed from the adhesives. Thereafter, appropriate colors and patterns relative to the reference edge and in register with the tab area can be printed. Following each printing station, the solvents should be flashed. Once complete, the printed and adhesive coated, scored film is rewound.

The printed and adhesive coated, scored roll of film is unwound into sheet cutting equipment. The film is then sheet cut relative to the printing and adhesive, and stacked. The stacks of labels are shear cut in register with label graphics and adhesive coating into stacks of individual label blanks. Die cut stacks of label blanks into finished labels relative to printing and adhesive. Following die cut, shrink wrap or otherwise package stacks of finished labels. During the blow- or injection-molding operation, unwrap and place stack of finished labels in feeding magazine. Pick-and-place device with vacuum end effector picks finished labels from magazine and places them individually in open container mold. Small vacuum ports in mold hold finished labels in position. Molds close and parisons are extruded into

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molds. Parisons are inflated to fill mold cavities. Heat from parisons activate adhesives on finished labels to effect bonds. Hollow blown containers are then cooled and removed from molds with finished  
5 labels strategically positioned thereon.

Examples of plastic in-mold labels of the present invention will now be further illustrated with reference to the following examples.

10 Example I - Sheets or Film

An unstretched sheet or film free of mechanically produced microvoids and formed with a continuous olefin resin matrix which includes an effective amount of a particulate filler having pre-existing  
15 microvoids, wherein the microvoid particulate filler is somewhat uniformly dispersed throughout the continuous olefin resin matrix for providing non-mechanically produced microvoids in communication with a surface of the in-mold label for imparting write-  
20 ability and printability thereto, is produced by mixing one of the following two compositions into a homogeneous blend and shaping the blend into pellet form:

	<u>Composition I</u>	<u>By Weight</u>
25	High density polyethylene	About 62%
	Polystyrene	" 7%
	Calcium carbonate	" 7%
	Diatomaceous earth	" 19%
	Titanium dioxide	" 5%

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<u>Composition II</u>	<u>By Weight</u>
High density polyethylene	About 71%
Polystyrene	" 8%
Calcium carbonate	" 5%
Diatomaceous earth	" 10%
Titanium dioxide	" 6%

5                   Upon mixing one of the above recited Compo-  
sitions, and in particular Composition I, to form a  
homogeneous blend and shaping the blend into a pellet,  
the pellet is extruded with, for example, a conven-  
tional single screw extruder into a film or sheet  
10                   having a continuous olefin resin matrix and a thick-  
ness of between about 1 mil and about 10 mils, and  
preferably about 4 mils to about 6 mils. The  
resulting unstretched film or sheet exhibits excellent  
receptivity and retentivity for common writing mate-  
15                   rials, such as ink, pencil lead, paint and the like,  
when it is written or printed on the surface thereof.

#### Example II - In-Mold Labels

20                   In addition to extrusion, the sheet or film  
of Example I can be blown or cast, embossed, matte or  
flat. In a further operation, a suitable heat acti-  
vatable adhesive is applied or coated onto selected  
sections on one side of the film. The adhesive is  
such that the adhesion properties are preferentially  
25                   activated in a subsequent blow-molding or injection-  
molding operation during application of the in-mold  
label to a plastic container. On the same or opposite

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side of the sheet or film, suitable score lines are made and selected decorative patterns are applied to the appropriate sections normally by printing, to provide an in-mold label design and information required for the finished article, such as a bottle or container. The printing can be done by various methods including rotogravure, offset, flexographic and the like. Surface primers may be coated onto the film prior to printing to enhance printing. Protective coatings, such as a lacquer, may be applied on top of the printing to protect the printing from moisture, abrasion, etc. The film is then die cut, used as label stock in an in-mold label application. In the case of offset printing, the adhesively coated, scored film may be first cut into sheets, then printed, and finally die cut into individual labels. For rotogravure printing, the sheet or film is printed and die cut after printing. After die cutting, the film is then in the proper shape and size for the final label.

The finished labels are applied to the bottle or container during the in-mold labeling operation. This process involves directly applying the in-mold labels via vacuum inside of the mold during the blow molding operation which forms the bottle or container. The semi-molten parison extruded into the mold when blown, expands and contacts the

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label strategically positioned within the mold and in particular the adhesive or release coating side thereof as well as the mold wall, at which point the molten plastic solidifies to form the finished  
5 article. The heat from the molten plastic activates the adhesive on the film and forms the bond between the bottle or container and the film.

The in-mold labels of this Example are believed to perform equally as well as equivalent  
10 paper-based products, with the added advantages of:  
1.) the in-mold labels may match the thermal characteristics, i.e., melting temperature, shrinkage, heat capacity, etc., of the blow molded plastic bottles or containers; and 2.) scrap molded bottles or containers  
15 formed with the in-mold labels of this Example may be easily recycled due to the fact that the in-mold labels are formed with materials similar in nature to those utilized to form the bottles or containers.  
Moreover, because the in-mold labels of this Example  
20 are formed with materials generally similar in nature to those utilized to form the bottles or containers, the conformity problem due to shrinkage normally associated with paper labels is substantially, if not  
25 performance during use of the bottles or containers is believed to be greatly enhanced by use of the in-mold labels of this invention.

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Thusly, it can be seen from the Examples, that the in-mold labels produced in accordance with the instant invention are effective substitutes for conventional pulp paper and the like. As a result, the in-mold labels of the instant invention provide simple, but unobvious means heretofore unavailable for providing in-mold labels.

The present invention may, of course, be carried out in other specific ways than those herein set forth without departing from the spirit and essential characteristics of the instant invention. The present embodiments are, therefore, to be considered in all respects as illustrative and not restrictive, and any changes coming within the meaning and equivalency range of the appended claims are to be embraced therein.

Having described our invention, we claim:



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1. A scored synthetic in-mold label for affixing to the outer surface of a substrate during the molding process of the substrate, said in-mold label comprising:

5 a plastic film layer having inner and outer surfaces;

a score line for dividing said plastic film layer into first and second sections; and

10 permanent bonding means, the inner surface of the first section of said plastic film layer having said permanent bonding means for permanently bonding the first section to the outer surface of the substrate, so that once said scored synthetic in-mold label has been affixed to the outer surface of the substrate, the second section of said plastic film layer may be separated from the first section along  
15 said score line to allow said second section to be removed from the substrate.

2. A scored synthetic in-mold label of claim 1, the inner surface of the second section having removable bonding means for removably bonding the second section to the outer surface of the substrate.

3. A scored synthetic in-mold label of claim 1, the outer surface of said plastic film layer having graphics printed thereon.

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4. A scored synthetic in-mold label of claim 1, the inner surface of said plastic film layer having graphics printed thereon.
5. A scored synthetic in-mold label of claim 1 wherein said plastic film layer has a thickness in the range of between about 1.0 mil and about 10 mils.
6. A scored synthetic in-mold label of claim 1 wherein said score line has a depth of from about 10% to about 50% of the thickness of said plastic film layer.
7. A scored synthetic in-mold label of claim 1 wherein said score line has a depth of about 33% of the thickness of said plastic film layer.
8. A scored synthetic in-mold label of claim 1, said permanent bonding means being a permanent adhesive.
9. A scored synthetic in-mold label of claim 2, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

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10. A scored synthetic in-mold label of claim 1, said plastic film layer being in the form of a multi-layer structure comprising a top layer and a bottom layer, said top layer having an outer surface with  
5 graphics printed thereon and the first section of said bottom layer having said permanent bonding means on its inner surface for permanently bonding the first section to the outer surface of the substrate.

11. A scored synthetic in-mold label of claim 10, said permanent bonding means being a permanent adhesive.

12. A scored synthetic in-mold label of claim 10, the second section of said bottom layer having removable bonding means on its inner surface for removably bonding the second section to the outer  
5 surface of the substrate.

13. A scored synthetic in-mold label of claim 12, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

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14. A scored synthetic in-mold label of claim 1, said plastic film layer being in the form of a multi-layer structure comprising a top layer and a bottom layer, said top layer having an inner surface with graphics printed thereon and the first section of said bottom layer having permanent bonding means on its inner surface for permanently bonding the first section to the outer surface of the substrate.

15. A scored synthetic in-mold label of claim 14, said permanent bonding means being a permanent adhesive.

16. A scored synthetic in-mold label of claim 14, the second section of said bottom layer having removable bonding means on its inner surface for removably bonding the second section to the outer surface of the substrate.

17. A scored synthetic in-mold label of claim 16, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

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18. A scored synthetic in-mold label of claim 1, said second section further having an edge tab for facilitating the separation of the second section from the first section along said score line and the  
5 removal of the second section from the substrate.

19. A scored synthetic in-mold label of claim 1, the substrate being a hollow plastic container produced by a process consisting of a blow-molding process and an injection-molding process.

20. A scored synthetic in-mold label of claim 1, said score line being on the inner surface of said plastic film layer.

21. A scored synthetic in-mold label of claim 1, said score line being on the outer surface of said plastic film layer.

22. A scored synthetic in-mold label of claim 1, the second section being in a form of a proof-of-purchase coupon.

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23. A scored synthetic in-mold label of claim 1, said plastic film layer having a second score line for dividing a third section of said plastic film layer from the second section, said third section having permanent bonding means on its inner surface for permanently bonding the third section to the outer surface of the substrate, so that once said scored in-mold label has been affixed to the substrate, the second section of said plastic film layer may be separated from the first section along said score line and from the third section along said second score line to allow said second section to be removed from the substrate.

24. A scored synthetic in-mold label of claim 23, said permanent bonding means being a permanent adhesive.

25. A scored synthetic in-mold label of claim 23, the third section having removable bonding means on its inner surface for removably bonding the third section to the outer surface of the substrate.

26. A scored synthetic in-mold label of claim 25, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

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27. A scored synthetic in-mold label of claim 23, said second section further having an edge tab for facilitating the separation of the second section from the first section along said score line and the third  
5 section along said second score line to allow the second section to be removed from the substrate.

28. A scored synthetic in-mold label of claim 1, said plastic film layer being substantially free of mechanically produced microvoids for writing and printing thereon, said plastic film layer further  
5 being formed of a continuous olefin resin matrix containing an effective amount of a particulate filler having microvoids to provide microvoids in communication with the outer surface of the plastic film layer for imparting writeability and printability thereto.

29. A scored synthetic in-mold label of claim 28 wherein said olefin resin is a polyethylene.

30. A scored synthetic in-mold label of claim 29 wherein said polyethylene is a high density polyethylene.

31. A scored synthetic in-mold label of claim 28 wherein said olefin resin is a polypropylene.

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32. A scored synthetic in-mold label of claim 28 wherein said particulate filler is a diatomaceous earth.

33. A scored synthetic in-mold label of claim 28 wherein said olefin resin is present in said continuous matrix in an amount of about 60% to about 90% by weight and said particulate filler is present in said  
5 continuous matrix in an amount of about 10% to about 40% by weight.

34. A scored synthetic in-mold label of claim 28 wherein said continuous olefin resin matrix further includes a styrene resin for imparting additional paper-like characteristics to said sectionalized  
5 synthetic in-mold label.

35. A scored synthetic in-mold label of claim 28 wherein said continuous olefin resin matrix further includes an inorganic filler.

36. A scored synthetic in-mold label of claim 28 wherein said inorganic filler is present in an amount of up to about 20% by weight.



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37. A scored synthetic in-mold label of claim 28 wherein said scored synthetic in-mold label has a thickness in the range of between about 1 mil and about 10 mils.

38. A scored synthetic in-mold label of claim 28 wherein said scored synthetic in-mold label has a thickness in the range of between about 4 mils and about 6 mils.

39. A scored synthetic in-mold label of claim 28 wherein said continuous olefin resin matrix further includes an anti-static agent.

40. A scored synthetic in-mold label of claim 28, said continuous olefin resin matrix comprising.

high density polyethylene,	about 60-75%
polystyrene,	about 5-10%
calcium carbonate,	about 5-10%
diatomaceous earth, and	about 10-20%, and
titanium dioxide	about 0-5%

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wherein said diatomaceous earth is dispersed throughout the continuous matrix to provide microvoids in communication with the outer surface of the plastic film layer to render said surface porous for imparting writeability and printability thereto.

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41. A scored synthetic in-mold label of claim 40, said continuous matrix comprising

high density polyethylene	about 62%,
polystyrene	" 7%,
calcium carbonate	" 7%,
diatomaceous earth	" 19%, and
titanium dioxide	" 5%.

42. A scored synthetic in-mold label of claim 40, said continuous matrix comprising

high density polyethylene	about 71%,
polystyrene	" 8%,
calcium carbonate	" 5%,
diatomaceous earth	" 10%, and
titanium dioxide	" 6%.

43. A scored synthetic in-mold label of claim 40, said continuous matrix further including an anti-static agent.

44. A scored synthetic in-mold label of claim 41, said continuous matrix further including an anti-static agent.

45. A scored synthetic in-mold label of claim 42, said continuous matrix further including an anti-static agent.

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46. A hollow plastic container having an outer sidewall surface and a scored synthetic in-mold label affixed to said outer sidewall surface during the molding process of said hollow plastic container, said scored synthetic in-mold label comprising a first section divided from a second section by a score line for permitting said second section to be separated from said first section and from the outer sidewall of said hollow plastic container along said score line.

47. A hollow plastic container of claim 46, said scored synthetic in-mold label comprising a plastic film layer having inner and outer surfaces, the inner surface of the first section of said plastic film layer having permanent bonding means for permanently bonding said first section to the outer sidewall surface of said hollow plastic container, so that said second section of said plastic film layer may be separated from said first section along said score line to allow said second section to be removed from said hollow plastic container.

48. A hollow plastic container of claim 46, said hollow plastic container being selected from a group of hollow plastic containers produced by a process consisting of a blow-molding process and an injection-molding process.

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49. A hollow plastic container of claim 47, said scored synthetic in-mold label having a second score line for dividing a third section from the second section, said third section having permanent bonding means on its inner surface for permanently bonding said third section to the outer sidewall of said hollow plastic container, so that said second section of said scored synthetic in-mold label may be separated from said first section along said score line and from said third section along said second score line to allow said second section to be removed from the outer sidewall of said hollow plastic container.

50. A hollow plastic container of claim 49, said second section having a removable adhesive on its inner surface for removably bonding said second section to the outer sidewall of said hollow plastic container, said first and third sections having a permanent adhesive on their inner surfaces for permanently bonding said first and third sections to the outer sidewall of said hollow plastic container.

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51. A hollow plastic container of claim 50, said second section further having an edge tab for facilitating the separation of said second section from said first section along said score line and from said third section along said second score line to allow said second section to be removed from said hollow plastic container.

52. A hollow plastic container of claim 47, said plastic film layer being in the form of a multi-layer structure comprising a top layer and a bottom layer, said top layer having an outer surface with graphics printed thereon and said first section of said bottom layer having said permanent bonding means on its inner surface for permanently bonding said first section to the outer sidewall of said hollow plastic container.

53. A hollow plastic container of claim 52, said permanent bonding means being a permanent adhesive.

54. A hollow plastic container of claim 52, said second section of said bottom layer having removable bonding means on its inner surface for removably bonding the second section to the outer sidewall of said hollow plastic container.

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55. A hollow plastic container of claim 54, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

56. A hollow plastic container of claim 47, said plastic film layer being in the form of a multi-layer structure comprising a top layer and a bottom layer; said top layer having an inner surface with graphics  
5 printed thereon and the first section of said bottom layer having permanent bonding means on its inner surface for permanently bonding said first section to the outer sidewall of said hollow plastic container.

57. A hollow plastic container of claim 56, said second section of said bottom layer having removable bonding means on its inner surface for removably  
5 bonding said second section to the outer sidewall of said hollow plastic container.

58. A hollow plastic container of claim 47, said permanent bonding means being a permanent adhesive.

59. A hollow plastic container of claim 57, said removable bonding means being selected from a group consisting of a removable adhesive and a release coating.

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60. A hollow plastic container of claim 47, said plastic film layer being substantially free of mechanically produced microvoids for writing and printing thereon, said plastic film layer further  
5 being formed of a continuous olefin resin matrix containing an effective amount of a particulate filler having microvoids to provide microvoids in communication with the outer surface of the plastic film layer for imparting writeability and printability thereto.

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61. In combination, a molded plastic substrate having an outer sidewall and a sectionalized synthetic in-mold label affixed to said outer sidewall during the molding operation of said molded plastic substrate, said sectionalized synthetic in-mold label comprising permanent and removable sections, the permanent section being divided from the removable section via a line of weakness for permitting the removable section to be separated from the permanent section and from the outer sidewall of said plastic substrate along said line of weakness by hand.

62. A combination of claim 61 wherein said molded plastic substrate is a hollow plastic blow-molded container.

63. A combination of claim 62 wherein said sectionalized synthetic in-mold label is in the form of a plastic film layer having inner and outer surfaces.



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64. A combination of claim 63, said plastic film layer being substantially free of mechanically produced microvoids for writing and printing thereon, said plastic film layer further being formed of a continuous olefin resin matrix containing an effective amount of a particulate filler having microvoids to provide microvoids in communication with the outer surface of the plastic film layer for imparting writeability and printability thereto.

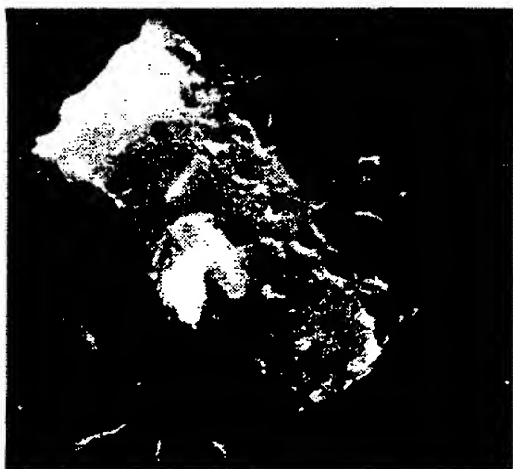
5



**FIG. 1**



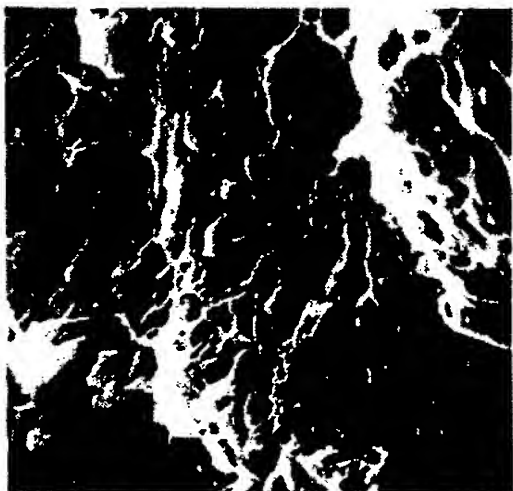
**FIG. 4**



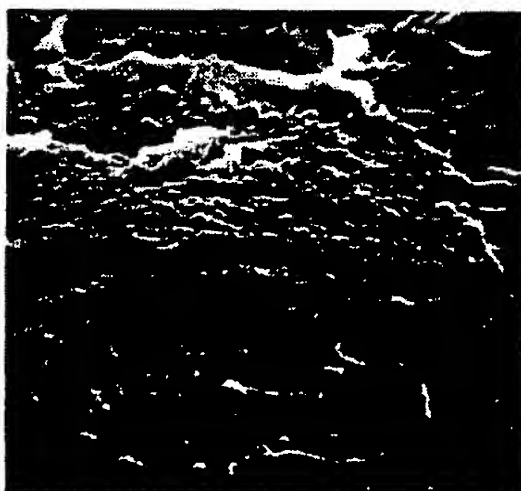
**FIG. 2**



**FIG. 5**



**FIG. 3**



**FIG. 6**

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FIG. 7

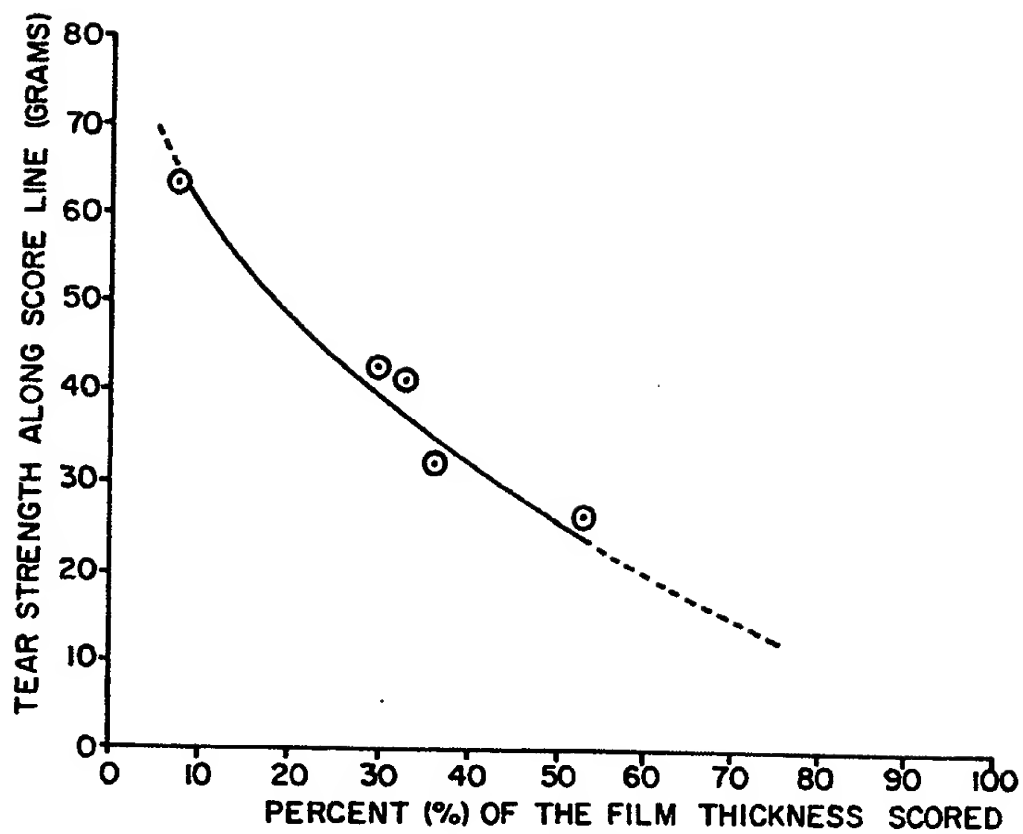


FIG. 8

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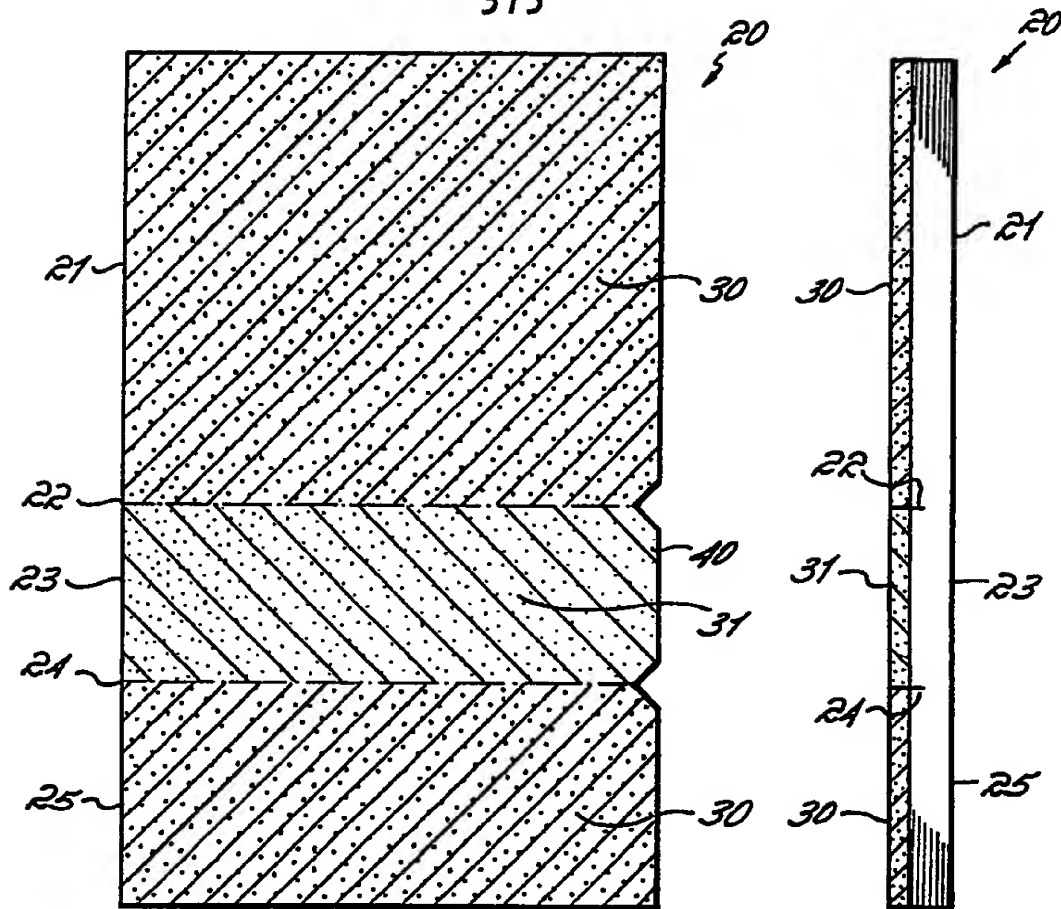


FIG. 9

FIG. 10

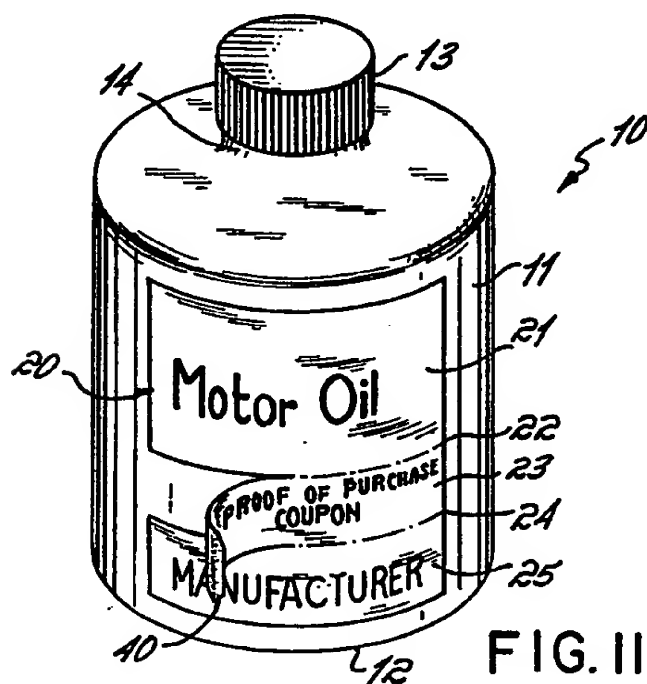


FIG. 11

<b>I. CLASSIFICATION OF SUBJECT MATTER</b> (If several classification symbols apply, indicate all) <sup>6</sup>		
According to International Patent Classification (IPC) or to both National Classification and IPC		
Int.Cl. 5 G09F3/04; . G09F3/02; B65D25/36		
<b>II. FIELDS SEARCHED</b>		
Minimum Documentation Searched <sup>7</sup>		
Classification System	Classification Symbols	
Int.Cl. 5	G09F ; B29C ; B65D	
Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in the Fields Searched <sup>8</sup>		
<b>III. DOCUMENTS CONSIDERED TO BE RELEVANT<sup>9</sup></b>		
Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
A	GB,A,2 119 746 (ET. MAURIUS MILLET) 23 November 1983 see page 1, line 63 - page 2, line 27; figures 1-5	1,46,61
A	FR,A,2 602 487 (VERRERIES GENERALES) 12 February 1988 see claims 1-10; figure 1	1,46,61
A	EP,A,0 349 670 (HAGEN AND SORENSEN) 10 January 1990	
<p><sup>10</sup> Special categories of cited documents:</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier document but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>"A" document member of the same patent family</p>		
<b>IV. CERTIFICATION</b>		
Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report	
02 MARCH 1992	04.03.92	
International Searching Authority	Signature of Authorized Officer	
EUROPEAN PATENT OFFICE	GALLO G.G. <i>G. Gallo</i>	

**ANNEX TO THE INTERNATIONAL SEARCH REPORT  
ON INTERNATIONAL PATENT APPLICATION NO. US 9107386  
SA 53792**

This annex lists the patent family members relating to the patent documents cited in the above-mentioned international search report.  
The members are as contained in the European Patent Office EDP file on  
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